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14. ABSTRACT Substantial technical progress was achieved during the reporting period in the following project areas:(A) increased organic-based materials options for materials with high permeability near room temperature (RT);(B) successfully initiated a program to prepare RT organic-based magnets that remain air stable to significantly above RT;(C) elucidated novel means for photonic control of magnetism through incorporation of photosensitive azo groups in the organic moiety;(D) Elucidated origins of magnetism and conductivity in RT organic-based magnet V[TCNE] <sub>x</sub> ;(E) Demonstrated the ability of organic semiconductors to transport spin in giant magnetoresistance(GMR)and tunneling magnetoresistance(TMR)device structures using rubrene as a model organic semiconductor;(F) demonstrated the ability of V(TCNE) <sub>x</sub> to inject spin into organic semiconductors such as rubrene, demonstrating the potential for all organic, flexible, air stable, light weight spintronics, as well as hybrid spintronics.					
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**FINAL PERFORMANCE REPORT**  
**March 1, 2006 to February 28, 2009**

**AFOSR Grant FA9550-06-1-0175**

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**ABSTRACT:**

Substantial technical progress was achieved during the reporting period in the following project areas: (A) increased organic-based materials options for materials with high permeability near room temperature (RT); (B) successfully initiated a program to prepare RT organic-based magnets that remain air stable to significantly above RT; (C) elucidated novel means for photonic control of magnetism through incorporation of photosensitive azo groups in the organic moiety; (D) Elucidated origins of magnetism and conductivity in RT organic-based magnet  $V[TCNE]_x$ ; (E) Demonstrated the ability of organic semiconductors to transport spin in giant magnetoresistance (GMR) and tunneling magnetoresistance (TMR) device structures using rubrene as a model organic semiconductor; (F) demonstrated the ability of  $V[TCNE]_x$  to inject spin into organic semiconductors such as rubrene, demonstrating the potential for all organic, flexible, air stable, light weight spintronics, as well as hybrid spintronics.

**I. OBJECTIVES**

Our goal, as stated in our 2005 proposal is “to establish the electromagnetic properties of organic-based magnets, and the electromagnetic properties of multilayer structures and composites, where one or more components are organic-based magnets.” For Years 1 and 2 of this program this was realized through synthesis, processing and characterization of photonically controlled organic-based magnets. By Year 3 we had greater understanding of the origins of the magnetic and charge transport properties of the room temperature magnet  $V[TCNE]_x$ . This led to realization of multilayer organic-based structures that operate as spintronic devices. It is anticipated that future work will further extend these materials and photonic and spintronic device concepts with potential for commercialization and defense and civilian use

**IV. ACCOMPLISHMENTS/NEW FINDINGS**

**YEAR 1 HIGHLIGHTS (details in Year 1 report):** We successfully developed (A) additional organic-based materials options for materials with high permeability at and above room temperature, (B) elucidated a novel mechanism for photoinduced change in magnetic permeability based on reversible photoinduced changes in magnetic anisotropy thereby enabling a potential means of photonic control of permeability and permittivity, (C) demonstrating the presence of multiple photonic responses in some organic-based magnets; (D) initial development of compositions more suitable for blending of magnetic and dielectric polymers to needed compositions of permittivity and permeability. Year 1 results reported in detail included:

**A. New room temperature magnets of  $V^{II}[TCNB]_{1.5}[TCNB]_x^{0.5}[V^{I}(CO)_6]_{1/2} \cdot zS$  (TCNB = tetracyanobenzene;  $4 > x > 3.5$ ;  $S = CH_2Cl_2, PhCF_3$ ) discovered.**

A new room temperature magnet of  $V[MeTCNC]_2$  was also discovered. MeTCNC is distinct from TCNE due to the presence of the  $O_2CMe$  group, which bonds to the V. New polymeric magnets are expected as the CH groups in both TCNB and MeTCNC can be substituted with polymer-compatible longer side chains.

**B. Novel mechanism of photoinduced magnetism in  $V(TCNE)_x$  elucidated**

During the first period, we discovered the photoinduced magnetic phenomena in room temperature organic-based magnet  $V(TCNE)_x$  [Publications 10 and 13 in Section VI]. The magnetization shows persistent and thermally reversible change induced by the  $\pi \rightarrow \pi^*$  excitation in  $(TCNE)^-$  anions. At low field, the magnetization of  $V(TCNE)_x$  decreases substantially by the illumination, whereas the high field magnetization remains the same as that of the ground state. The light-induced IR spectra suggest an activation to a metastable state, which should be preserved for long period time at low temperature ( $T < 100$  K).

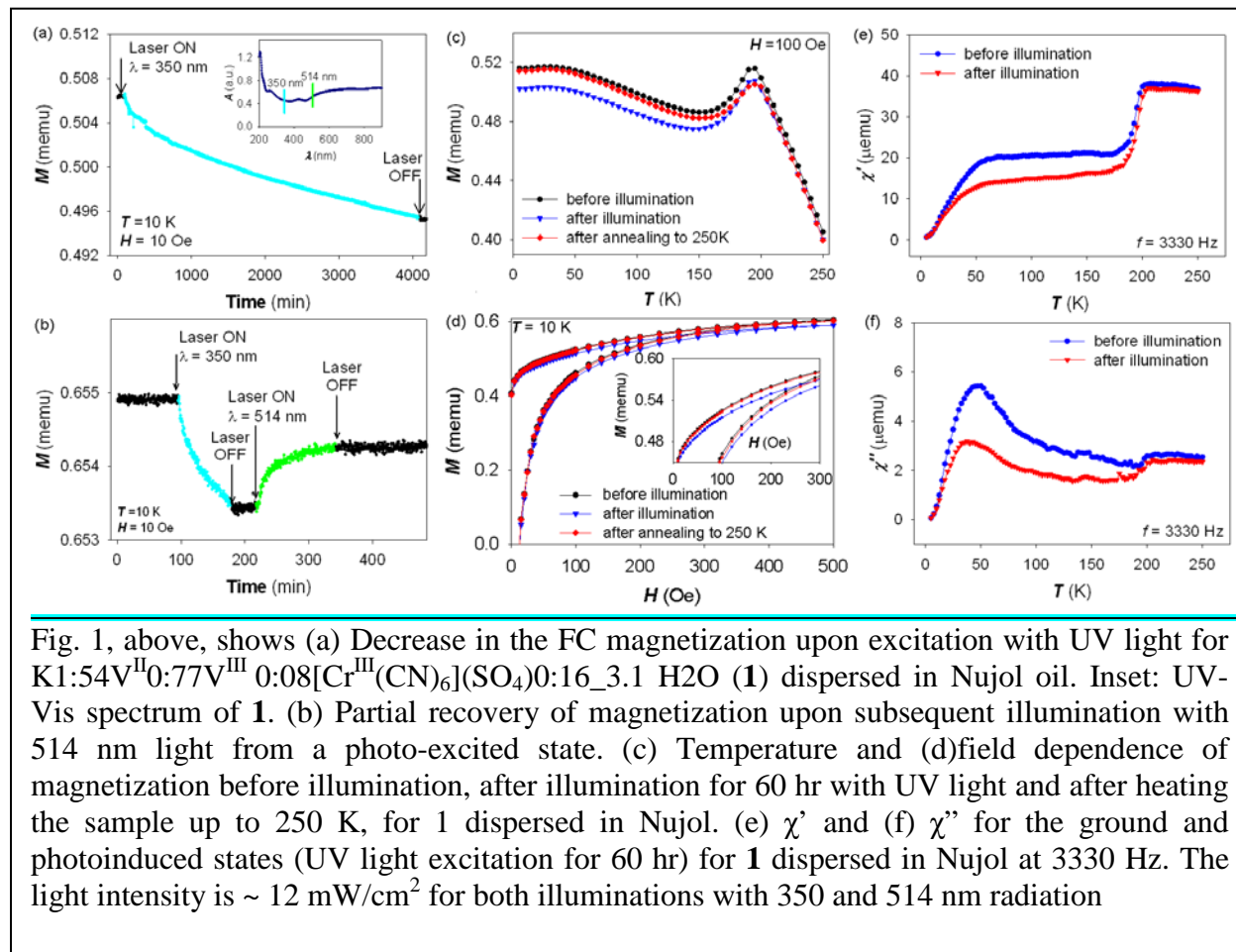
**YEAR 2 HIGHLIGHTS (details in Year 2 report):**

**A. Synthesis and Control of Processing:**

During Year 2 we have concentrated our physical studies on  $V[TCNE]_x$ , the first reported molecule-based magnets that is magnetic to above room temperature (to 420 K), and  $V_x[Cr(CN)_6]_y \cdot zH_2O$  Prussian Blue, a second molecule-based magnet that can be magnetic to above room temperature. We also have carried out extensive studies of the chemistries related to the synthesis and processing of these two classes of materials and other promising classes of molecule-based magnetic materials. The chemistries studied are largely presented in the published papers 1 – 6 listed in Section VI below.

**B. Optical Control of Magnetization in The Room Temperature Magnet: V-Cr Prussian Blue Analog**

Despite a long period of intense worldwide research on photoinduced magnetism, the commercial application is yet to be realized. One of the major challenges is to design and develop new materials that exhibit both photoinduced magnetism and long-range magnetic order at higher temperature. Our search for new light-tunable magnets brought about a new discovery of reversible photoinduced magnetism in the system that exhibits magnetic order well above room temperature. A Prussian blue magnet analog of  $V_x[Cr(CN)_6]_y \cdot zH_2O$  is one of the few



molecule-based room temperature magnets. It also is a good candidate for the application of inexpensive and large area terahertz imaging technology due to its relatively high permeability. We have built on the progress we reported last year in the synthesis of a mixed valent Vanadium Hexacyanochromate with a  $T_c$  around 372 K. We report reversible photoinduced magnetic phenomena for a V-Cr Prussian blue analog ( $T_c \sim 350 \text{ K}$ ). The molecule-based magnet exhibits a decrease in magnetization upon illumination with UV light ( $\lambda \sim 350 \text{ nm}$ ) and reaches a metastable state that has a long lifetime at low temperatures ( $> 10^6 \text{ s}$  at 10 K). This photo-excited magnetic state totally recovers back to the ground state by warming above 250 K, and partially recovers with green light ( $\lambda \sim 514 \text{ nm}$ ) illumination. The effect of green light is triggered only when the sample is previously UV-irradiated, suggesting a hidden metastable magnetic state. The photoinduced magnetic effects are proposed to originate from structural distortion that alters the magnetic exchange coupling and/or anisotropy. The research on photoinduced magnetism (PIM) has received broad attention not only for its novel phenomena but more importantly on its

possible application in light controlled antennas and detectors and also the possibility of light controlled memory devices. Publication 47 listed in Section VI presents our more recent results describing sample preparation, experimental techniques and result, and a discussion concerning the likely mechanisms operating. Fig. 1, above shows (a) Decrease in the FC magnetization upon excitation with UV light for  $\text{K1:54V}^{\text{II}}\text{0:77V}^{\text{III}}\text{0:08}[\text{Cr}^{\text{III}}(\text{CN})_6](\text{SO}_4)\text{0:16}_3\cdot 1\text{ H}_2\text{O}$  (**1**) dispersed in Nujol oil. Inset: UV-Vis spectrum of **1**. (b) Partial recovery of magnetization upon subsequent illumination with 514 nm light from a photo-excited state. (c) Temperature and (d) field dependence of magnetization before illumination, after illumination for 60 hr with UV light and after heating the sample up to 250 K, for **1** dispersed in Nujol. (e)  $\chi'$  and (f)  $\chi''$  for the ground and photoinduced states (UV light excitation for 60 hr) for **1** dispersed in Nujol at 3330 Hz. The light intensity is  $\sim 12\text{ mW/cm}^2$  for both illuminations with 350 and 514 nm radiation.

### **C. Irreversible Magnetic Bistability and Nucleation of Magnetic Bubbles in Layered 2D Organic-based Magnet $[\text{Fe}(\text{TCNE})(\text{NCMe})_2][\text{FeCl}_4]$**

The 2D layered organic-based magnet  $[\text{Fe}(\text{TCNE})(\text{NCMe})_2][\text{FeCl}_4]$  (TCNE  $\equiv$  tetracyanoethylene) exhibits a unique magnetic bifurcation between the field-cooled and zero-field-cooled states, which cannot be explained by either superparamagnetic behavior or spin freezing due to spin glass order. This irreversible magnetic bistability is described through consideration of the ensemble of uncoupled 2D Ising layers and their magnetization reversal initiated by a field-induced bubble formation (*bubble bursting*) in individual layers. The bubble nucleation rate strongly depends on the external field and temperature resulting in anomalous magnetic relaxation. Publication 33 listed in Section VI provides detailed results and a detailed description of the model for formation and bursting of the spin bubbles.

Recently developed  $[\text{Fe}(\text{TCNE})(\text{NCMe})_2][\text{FeCl}_4]$  has the first 2D layered structure determined in this family of magnets. The structure consists of undulating layers composed of  $\text{Fe}^{\text{II}}$  ions with  $\mu_4$ - $[\text{TCNE}]^{\bullet -}$  bridging within the layer and two axial MeCNs coordination. There are no covalent bonds between layers. Additional paramagnetic  $[\text{FeCl}_4]^-$  anions occupy sites between the layers of  $[\text{Fe}(\text{TCNE})(\text{NCMe})_2]^+$  but do not contribute to the magnetic ordering. The absence of bridging ligands between the layers suggests only dipolar coupling exists between the layers. In each layer, the magnetic coupling between spin in  $[\text{TCNE}]^{\bullet -}$  ( $S = 1/2$ ) and spins in  $\text{Fe}^{\text{II}}$  ( $S = 2$ ) is suggested to be antiferromagnetic resulting in ferrimagnetic order similar to other metal-TCNE magnets. Thus, each individual layer is considered as an ideal 2D Ising system. Publication 19 in Section VI gives a detailed report of our solution for the crystal structure of this model metal-TCNE material.

### **D. Bonding, Backbonding, and Spin-Polarized Molecular Orbitals: Basis for Magnetism and Semiconducting Transport in $\text{V}[\text{TCNE}]_{x\sim 2}$**

X-ray absorption spectroscopy (XAS) and magnetic circular dichroism (MCD) at the V  $L_{2,3}$  and C and N  $K$  edges reveal bonding and backbonding interactions in films of the 400 K magnetic semiconductor  $\text{V}(\text{TCNE})_{x\sim 2}$ . In V spectra,  $d_{xy}$ -like orbitals are modeled assuming  $\text{V}^{2+}$  in an octahedral ligand field, while  $d_z^2$  and  $d_{x^2-y^2}$  orbitals involved in strong covalent  $\sigma$  bonding cannot be modeled by atomic calculations. C and N MCD, and differences in XAS from neutral TCNE

molecules, reveal spin-polarized molecular orbitals in  $V(\text{TCNE})_{x \sim 2}$  associated with weaker  $\pi$  bonding interactions that yield its novel properties. Further details are in publication 32 in Section VI

### **E. Detection of Weak and Strong Magnetic Fields Using Organic Semiconductors**

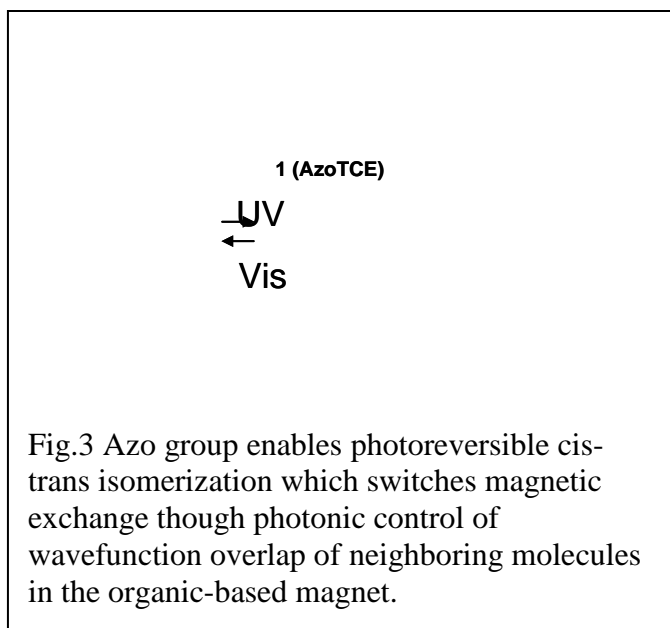
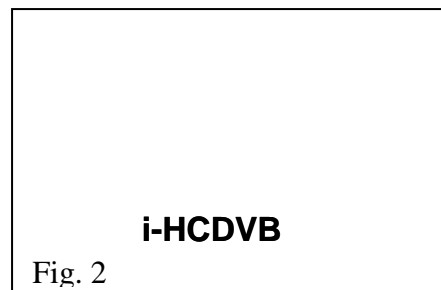
**Publications 29 (experimental results) and 40 (theoretical models we developed) in Section VI describe these phenomena.** We reported that organic semiconductors such as  $\alpha$ -sexithiophene ( $\alpha$ -6T) have magnetoresistance (MR) with unexpected sign changes; depending on applied voltage, temperature, and layer thickness, the resistance may either increase or decrease upon application of a small magnetic field (<100 mT). We proposed that MR and the inversion of MR are due to the role of hyperfine interaction in a magnetic field, as illustrated by the recombination-limited regime.

We determined that the recombination rate of coulombically bound electron-hole pairs depends on their spin configuration. Because of low spin-orbit coupling the spin dynamics of these well separated coulombic pairs is determined by weak hyperfine and exchange interactions. In this case a weak magnetic field produces a strong effect on the spin dynamics and hence on the recombination rate of e-h pairs. We have shown that the recombination current in organic semiconductors may have a maximum as a function of recombination constant. For high recombination constants the current is space charge limited and decreases with increasing the e-h recombination constant. This decay of current is due to decrease of the region where the recombination takes place. At a low recombination constant the recombination takes place in the whole volume and the current increases with increasing the recombination constant. The characteristic recombination constant separating those two regimes depends on the thickness of sample, applied voltage, and charge carrier mobilities. The model predictions are consistent with experimental data for magnetoresistance of organic semiconductors.

## **Year 3 – Selected Achievements**

### **A. New Approach to Photo-controllable Organic-based Magnets**

Several new  $V(\text{TCNE})$ -based organic-based magnets were synthesized by replacing TCNE with different organic molecules which also have numerous nitrile groups. the Tc's of these new magnets range from room temperature to low temperature. We proposed a new idea which takes advantage of the chemical tailorability of organic molecules to create photo-controllable organic magnets.



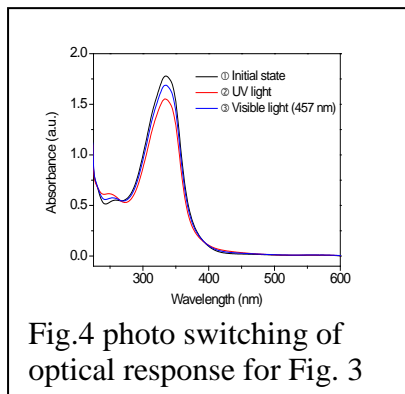


Fig.4 photo switching of optical response for Fig. 3

Several new electron acceptors are designed with azo ( $-N=N-$ ) group and three or six nitrile groups. The nitrile groups are expected to react with vanadium forming an organic-based magnet and the azo group which has the photoisomerization property under UV/vis light would play the role to tune the magnetic property through changing the molecular overlaps. Fig.4 shows the photo switching of the optical response of Fig.3. Photoswitching of magnetic response is still under investigation. Preliminary results at 10 K show photocontrolled decrease in magnetism of  $\sim 20\%$ , reversible by thermal annealing. Further synthesis and physical studies are in progress.

#### A. Giant magnetoresistance in ferromagnet/organic semiconductor/ferromagnet heterojunctions (see Publications 37 and 39 in Section 6 – note publication 37 is an “editor’s select pick in Physical Review B”)

We reported in the spin injection and transport in ferromagnet/organic semiconductor/ferromagnet (FM/OSC/FM) heterojunctions using rubrene ( $C_{42}H_{28}$ ) as an organic semiconductor spacer. For completeness of our study, both tunneling magnetoresistance (TMR) and giant magnetoresistance (GMR) were studied by varying the thickness of the rubrene layer (5–30 nm). A thorough study of the device characteristics reveals spin-polarized carrier injection into and subsequent transport through the OSC layer. When the thickness of the rubrene layers are beyond the tunneling limit, the device currents are limited by carrier injection and bulk transport. The carrier injection is well described with phonon-assisted field emission. The behavior of GMR in response to bias field and temperature shows significant differences from that of TMR. Fig. 5 is a typical curve showing operation of a FM/OSC/FM heterojunction using rubrene ( $C_{42}H_{28}$ ) as an organic semiconductor spacer. The results support the promise of use of organic semiconductors (OSCs) in light weight spintronic devices.

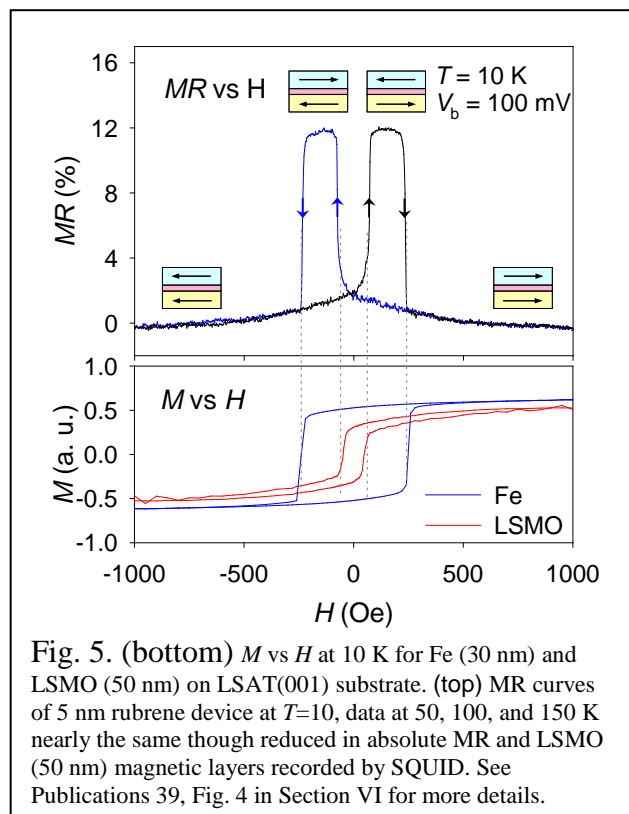
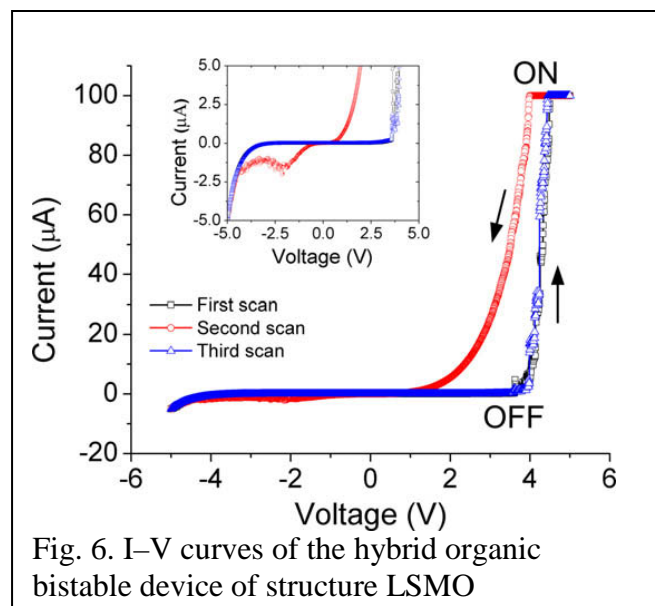


Fig. 5. (bottom)  $M$  vs  $H$  at 10 K for Fe (30 nm) and LSMO (50 nm) on LSAT(001) substrate. (top) MR curves of 5 nm rubrene device at  $T=10$ , data at 50, 100, and 150 K nearly the same though reduced in absolute MR and LSMO (50 nm) magnetic layers recorded by SQUID. See Publications 39, Fig. 4 in Section VI for more details.

### C. Electrical bistability and spin valve effect in a ferromagnet/organic semiconductor/ferromagnet heterojunction (for further information see publication 45 in Section VI)

We reported discovery and study of the electrical bistability and bias-controlled spin valve effect



in an organic spintronics device using rubrene ( $C_{42}H_{28}$ ) as an organic semiconductor channel. The half-metallic  $La_{0.7}Sr_{0.3}MnO_3$  (LSMO) and Fe are used as the two ferromagnetic electrodes. The device displays reproducible switching between a low-impedance (ON) state and a high impedance (OFF) state by applying different polarities of high biases. In the ON state, the device shows a spin valve effect with magnetoresistance values up to 3.75%. The observed spin valve effect disappears when the device recovers to the initial OFF state, Fig. 6. This opens the opportunity for multifunctional organic-based spintronic devices using both magnetic and electric fields to manipulate information.

### D. Spin injection/detection using an organic-based magnetic semiconductor

The new paradigm of electronics, ‘spintronics’, promises to extend the functionality of information storage and processing in conventional electronics. The principal spintronics device, the ‘spin valve’, consists of two magnetic layers decoupled by a spin-transporting spacer, which allows parallel (on) and antiparallel (off) alignment of the magnetizations (spins) of the two magnetic layers. The device resistance then depends on the spin alignment controlled by the external magnetic field. In pursuit of semiconductor spintronics there have been intensive efforts devoted to develop room temperature magnetic semiconductors and also to incorporate both inorganic semiconductors and carbon-based materials as the spin-transporting channels. Molecule/organic-based magnets, which allow chemical tuning of electronic and magnetic properties, are a promising new class of magnetic materials for future spintronic applications. We reported the first realization of an organic-based magnet as an electron spin polarizer in the standard spintronics device geometry. (Publication 44 in Section VI, Nature Materials, in press). A thin non-magnetic organic semiconductor layer and an epitaxial ferromagnetic oxide film were employed to form a hybrid magnetic tunnel junction. The results demonstrate the spin-polarizing nature of the organic-based magnetic semiconductor,  $V(TCNE)_x$  ( $x \sim 2$ ;  $T_c \sim 400$  K), and its function as a spin injector/detector in hybrid magnetic multilayer devices, Fig. 7.

Our success demonstrates the possibilities for “all-organic spintronics”, as well as “hybrid spintronics”, and “air stable, light-weight, flexible spintronics”. We have initiated studies of these extensions to organic spintronics, with promising preliminary results.



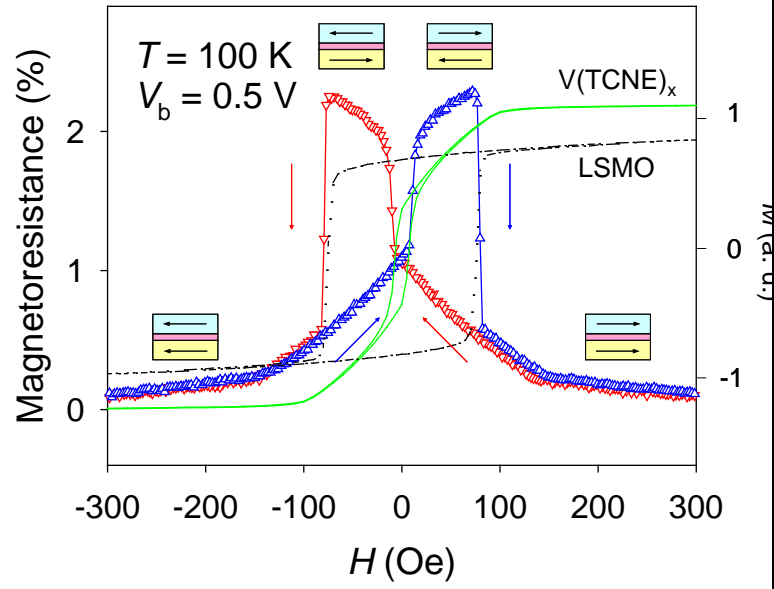


Fig. 7 The magnetoresistance curves of a  $V(\text{TCNE})_x(500 \text{ nm})=\text{rubrene}(5 \text{ nm})=\text{LAO}(1:2 \text{ nm})=\text{LSMO}(80 \text{ nm})$  junction measured at 100 K with a bias field of 0.5V. The blue (red) line and up triangles (down triangles)

## V. PERSONNEL SUPPORTED (03-01-2006 to 02-28-2009)

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35. J.S. Miller and K.S. Min, *Oxidation Leading to Reduction - Redox-Induced Electron Transfer (RIET)*, **Angewandte Chemie International Edition**, **48**, 262-272 (2009).
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37. J.-W. Yoo, H.W. Jang, V.N. Prigodin, C. Kao, C.B. Eom, and A.J. Epstein, *Giant Magnetoresistance in Ferromagnet/Organic Semiconductor/Ferromagnet Heterojunctions*, **Physical Review B** **80**, 205207/1-9 (2009).
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  42. M.B. Murphey, J.D. Bergeson, S.J. Etzkorn, L. Qu, L. Li, L. Dai, and A.J. Epstein, *Spin-valve Behavior in Porous Alumina-Embedded Carbon Nanotube Array with Cobalt Nanoparticle Spin Injectors*, **Synthetic Metals** **160**, 235-237 (2010).
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  47. K.D. Bozdag, J.-W. Yoo, N.P. Raju, A.C. McConnell, J.S. Miller, and A.J. Epstein, *Optical control of Magnetization in The Room Temperature Magnet: V-Cr Prussian Blue Analog*, submitted.

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Degree: Ph.D. Physics  
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## VII. INTERACTIONS/TRANSITIONS (03-01-2006 to 02-28-2009)

### VII. a. Participation/presentations at meetings, conferences, seminars, etc.

#### Invited Talks, Arthur J. Epstein:

1. A.J. Epstein, *Extraordinary Magnetotransport Phenomena in Organic Semiconductors*, **2nd International Workshop of NANO Systems Institute, Seoul National University**, Seoul, Korea, May 8, 2006.
2. A.J. Epstein, Keynote Lecture, *Organic-Based Magnets and Spintronics*, **The International Conference on Science and Technology of Synthetic Metals (ICSM) 2006**, Dublin, Ireland, July 2-7, 2006.
3. A.J. Epstein, Plenary Lecture, *Spin Glass Phenomena in Molecule-based Magnets*, **10th International Conference on Molecule-based Magnets (ICMM)**, Victoria, B.C., Canada, August 13-17, 2006.
4. A.J. Epstein, *Organic-Based Magnets and Spintronics*, **Symposium on Polymer Electronics and Nanoscience**, Linkoping, Sweden, August 18, 2006.
5. A.J. Epstein, *Multiple Photonic Responses in Organic-based Magnets*, **International Symposium on Biophotonics, Nanophotonics, and Metamaterials**, Hangzhou, PR China, October 16, 2006.
6. A.J. Epstein, *From Organic Conductors to Organic Magnets-Three Decades of Collaboration with a Chemist on New Physics and New Chemistry*, **Joint Symposium on Initiatives for Attractive Education in Graduate Schools of Physics and Chemistry** (and “Application of Advanced Analytical Techniques to Chemical Substances), Tokyo, Japan. February 2, 2007.
7. A.J. Epstein, *New Physics from New Magnetic Materials*, **Seminar of Prof. A. Epstein and Prof. G. Tatara as a Satellite of Joint Symposium on Initiatives for Attractive Education in Graduate Schools of Physics and Chemistry** (and Application of Advanced Analytical Techniques to Chemical Substances), Tokyo, Japan, February 3, 2007.
8. A.J. Epstein, *Predictable and New Physics and Potential for Applications of Organic-based Magnets*, **Meeting of the American Physical Society**, Denver, Colorado, March 5-9, 2007 [*James C. McGroddy Award Address*].



9. J.D. Bergeson and A.J. Epstein, *Low Field, Large Magnetoresistance in Nonmagnetic Organic Semiconductors*, **Meeting of the American Physical Society**, Denver, Colorado, March 5-9, 2007).
10. A.J. Epstein, *Surprising Very Large Impact of Small Magnetic and Electric Fields on Conducting Polymers*, **233<sup>th</sup> National Meeting of the American Chemical Society**, Chicago, Illinois, March 25-29, 2007.
11. A.J. Epstein, J.W. Yoo, R. Shima Edelstein, D.M. Lincoln, N.P. Raju, and J.-Y. Chen, *Optical Control of Random Magnetic Anisotropy in Organic-Based Magnetic Semiconductor  $V(TCNE)_x$ ,  $x \sim 2$  Film*, **7<sup>th</sup> International Conference on Optical Probes of  $\pi$ -Conjugated Polymers and Functional Self Assemblies (OP 2007)**, Turko, Finland, June 11-15, 2007.
12. A.J. Epstein, *New Physics in Organic-Based Magnets*, **Symposium on Organic Magnetism**, Oak Ridge, Tennessee, July 16, 2007.
13. A.J. Epstein, *Organic-based Magnetic Semiconductors and Potential for Spintronics*, **SpinOS 2007: Workshop on Spintronic Effects in Organic Semiconductors**, Bologna, Italy, September 9-11, 2007.
14. A.J. Epstein, *Magnetic and Nonmagnetic Organic Semiconductors and Magnetoresistance: Towards Organic-based Spintronics*, **7<sup>th</sup> International Symposium on Crystalline Organic Metals Superconductors and Ferromagnets**, Peñíscola, Spain, September 24-29, 2007.
15. A.J. Epstein, *Organic-Based Magnets and Magnetic Phenomena in Organics: New Science and New Technologies*, **International Conference on Science and Technology of Synthetic Metals**, Porto de Galinhas, Brazil, July 6-11, 2008.
16. A.J. Epstein, *New Prospects in Molecular-Based Magnets*, **The 57<sup>th</sup> Fujihara Seminar, International Conference on New Prospects on Molecular Magnetism**, Tomakomai, Hokkaido, Japan, July 28-31, 2008.
17. A.J. Epstein, *New Magnetic Phenomena Enabled by 'Hybrid' Organic-based Magnets*, **G-COE International Symposium: Perspectives in Inorganic-Organic Hybrid Materials**, Sendai, Japan, August 1, 2008.
18. A.J. Epstein, *Opportunities in Extended Organic-based Magnet Materials: From Fractal Magnets to Photonically Controlled Magnetism and Organic-based Magnetic Semiconductors*, **2<sup>nd</sup> Workshop on "Current Trends in Nanoscopic and Mesoscopic Magnetism"**, Delphi, Greece, September 2-5, 2008.
19. A.J. Epstein, J.-W. Yoo, V.N. Prigodin, W.W. Shum, K.I. Pokhodnya, J.S. Miller, *Irreversible Magnetic Bistability and Nucleation of Magnetic Bubbles in Layered 2D*

*Organic-based Magnet [Fe(TCNE)(NCMe)<sub>2</sub>][FeCl<sub>4</sub>], 11th International Conference on Molecule-based Magnets (ICMM 2008) Florence, Italy, September 21-24, 2008.*

20. A.J. Epstein, *Organic Based Magnets and Magnetic Phenomena in Organics*, **7<sup>th</sup> KCIST (Korea Conference on Innovative Science and Technology) Electronic Properties of Carbon-based Materials**, Phoenix Park Korea, October 19, 2008.
21. A.J. Epstein, *Magnetotransport in Organic Semiconductors*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
22. V.N. Prigodin and A.J. Epstein, *Spin Dynamics Control of Recombination Current in Organic Semiconductors*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
23. A.J. Epstein, *Organic Magnetoresistance*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009.
24. Y.W. Yoo, V.N. Prigodin, A.J. Epstein, *Photoinduced Magnetism and Spintronics in Organic Semiconductors*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009.
25. A.J. Epstein, Plenary Lecture, *Spins in Organic Semiconductors: New Physics and New Technologies*, **55<sup>th</sup> Midwest Solid State Conference, University of Iowa**, Iowa City, IA, April 18-19, 2009.
26. A.J. Epstein, *Molecular/Organic based Magnetism: New Chemistry, Physics, and Technologies*, **AVS 56<sup>th</sup> International Symposium and Exhibit, American Vacuum Society Meeting, San Jose, California**, November 9-13, 2009.
27. J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein, *Moving toward Organic Spintronics*, **International Symposium on Carbon Electronics 2010, Seoul National University**, Seoul, Korea, May 3, 2010.
28. V.N. Prigodin, C.-Y. Chen, J.W. Yoo, C.B. Eom, H.W. Jang, C.W. Bark, and A.J. Epstein, *Spin Injection/Detection with Molecule-based Magnets: New Science and Opportunities*, **Trends in Spintronics and Nanomagnetism (TSN-2010)**, Lecce, Italy, May 23-27, 2010.
29. A.J. Epstein, V.N. Prigodin, J.-W. Yoo, D. Bozdog, C.-Y. Chen, L. Fang, and E. Johnston-Halperin, *Advances in Organic-based Magnets and Organic-based Spintronics*, **ISSP-MDF Joint International Workshop: Spin-related Phenomena in Organic Materials**, Kashiwa, Chiba, Japan, July 1-3, 2010.

30. A.J. Epstein, Keynote Lecture, Organic-Based Magnets and Organic-Based Spintronics, **International Conference on Science and Technology of Synthetic Metals (ICSM 2010)**, Kyoto, Japan, July 4-9, 2010.

Contributed Talks, Arthur J. Epstein

1. J.W. Yoo, R. Shima Edelstein, A.J. Epstein, K. Pokhodnya, and J.S. Miller, *Photoinduced Magnetic and Electronic Phenomena in Organic Magnetic Semiconductor  $V(TCNE)_{x-2}$* , **Meeting of the American Physical Society**, Baltimore, Maryland, March 13-17, 2006 [Bulletin of the American Physical Society 51, 628 (2006)].
2. J. Kortright, R. Shima Edelstein, D.M. Lincoln, J.W. Yoo, and A.J. Epstein, *V, C, and N soft x-ray absorption and MCD of molecular magnet  $V(TCNE)_{x-2}$  films*, **Meeting of the American Physical Society**, Baltimore, Maryland, March 13-17, 2006 [Bulletin of the American Physical Society 51, 1515 (2006)].
3. J.D. Bergeson, D. M. Lincoln, R. Shima Edelstein, V. Prigodin, and A.J. Epstein, *Anomalous Magnetoresistance Phenomena in Organic Semiconductors*, **Meeting of the American Physical Society**, Baltimore, Maryland, March 13-17, 2006 [Bulletin of the American Physical Society 51, 1525 (2006)].
4. R. Shima Edelstein, D. M. Lincoln, J.W. Yoo, N.P. Raju, J.D. Bergeson, and A.J. Epstein, *Preparation, Magnetism, and Applications of Thin Films of the Organic Semiconductor  $V(TCNE)_{x-2}$* , **Meeting of the American Physical Society**, Baltimore, Maryland, March 13-17, 2006 [Bulletin of the American Physical Society 51, 1525 (2006)].
5. N.P. Raju, R. Shima Edelstein, and A.J. Epstein, *Magnetoresistance and Ferrimagnetic Resonance (FMR) on Thin Films of the Organic-based Magnetic Semiconductor  $V(TCNE)_{x-2}$  with  $T_c$  above 350 K.*, **Meeting of the American Physical Society**, Baltimore, Maryland, March 13-17, 2006 [Bulletin of the American Physical Society 51, 1525 (2006)].
6. Y. Bataiev, N.P. Raju, K.I. Pokhodnya, J.S. Miller, and A.J. Epstein, Nanoscale Control of  $V_xCo_{1-x}[TCNE]_2$  Organic-Based Magnetic Semiconductors, **The Ohio Nanotechnology Summit**, Columbus, Ohio, April 4-5, 2006.
7. J.D. Bergeson, D.M. Lincoln, V.N. Prigodin, and A.J. Epstein, *Anomalous Room Temperature Magnetoresistance in Thin-Film Organic Semiconductor Devices*, **The Ohio Nanotechnology Summit**, Columbus, Ohio, April 4-5, 2006.
8. R. Shima Edelstein, D.M. Lincoln, Jung-Woo Yoo, N.P. Raju, J.D. Bergeson, J.B. Kortright, and A. J. Epstein, *Nanoscale Control of Preparation, Magnetism, and Applications of Thin Films of the Organic Semiconductor  $V[TCNE]_{x-2}$* , **The Ohio Nanotechnology Summit**, Columbus, Ohio, April 4-5, 2006.

9. K. Pokhodnya, M. Bonner, J. Her, P. Stephens, J. Miller, and A.J. Epstein, *Magnetic Ordering of New Molecule-Based Magnet:  $[Fe(TCNE)(NCMe)] [FeCl_4]$* , **Meeting of the American Physical Society**, Denver, Colorado, March 5-9, 2007 (<http://meetings.aps.org/link/BAPS.2007.MAR.J16.13>)
10. J.W. Yoo, R. Shima Edelstein, D.M. Lincoln, and A.J. Epstein, *Ferrimagnetic Resonance Study on Photo-induced Magnetism in Hybrid Magnetic Semiconductor  $V(TCNE)_x$ ,  $x \sim 2$  Film*, **Meeting of the American Physical Society**, Denver, Colorado, March 5-9, 2007 (<http://meetings.aps.org/link/BAPS.2007.MAR.V25.6>).
11. N. Raju, K. Pokhodnya, J. van Tol, J. Miller, and A.J. Epstein, *High Frequency (240 GHz) Ferrimagnetic Resonance (FMR) of Room Temperature Organic Based Magnetic Semi-Conductor  $V[TCNE] (x \sim 2)$* , **Meeting of the American Physical Society**, Denver, Colorado, March 5-9, 2007 (<http://meetings.aps.org/link/BAPS.2007.MAR.J16.9>)
12. J.W. Yoo, R. Shima Edelstein, D.M. Lincoln, N. Raju, K. Pokhodnya, J.S. Miller and A.J. Epstein, *Novel Mechanism of Photoinduced Change in Magnetism and Conductivity in Organic-based Magnetic Semiconductor,  $V(TCNE)_x$ ,  $x \sim 2$* , **52<sup>nd</sup> Annual Conference on Magnetism and Magnetic Materials**, Tampa, Florida, November 5-9, 2007 [Abstract HD-11].
13. J.B. Kortright, D.M. Lincoln, R. Shima Edelstein, and A.J. Epstein, *Element -Resolved Magnetism in Organic-Based Magnetic Semiconductor  $V(TCNE)_{x \sim 2}$* , **52<sup>nd</sup> Annual Conference on Magnetism and Magnetic Materials**, Tampa, Florida, November 5-9, 2007 [Abstract HD-08].
14. J.-W. Yoo, V.N. Prigodin, K.I. Pokhodnya, J.S. Miller, and A.J. Epstein, *Anomalous Magnetic Relaxation in 2D Layered Organic-Based Magnet  $[Fe(TCNE)(NCMe)_2] [FeCl_4]$* , **Meeting of the American Physical Society**, New Orleans, Louisiana, March 10-14, 2008 (<http://meetings.aps.org/Meeting/MAR08/Event/75495>).
15. N.P. Raju, J.W. Yoo, A.C. McConnell, W.W. Shum, K.J. Nelson, J.S. Miller, and A.J. Epstein, *Ferromagnetic Resonance (FMR) of A Room Temperature Organic Based Mixed Valent Vanadium Hexacyanochromate Magnet  $K_{1.54}V^{II}_{0.77}V^{II}_{0.08} [Cr^{III}(CN)_6](SO_4)_{0.16} \cdot 3.1H_2O$* , **Meeting of the American Physical Society**, New Orleans, Louisiana, March 10-14, 2008 (<http://meetings.aps.org/Meeting/MAR08/Event/75496>).
16. K.D. Duman, J.-W. Woo, N.P. Raju, A.C. McConnell, W.W. Shum, K.J. Nelson, J.S. Miller, and A.J. Epstein, *Reversible Photoinduced Magnetism in V-Cr Prussian Blue Analogues*, **Meeting of the American Physical Society**, New Orleans, Louisiana, March 10-14, 2008 (<http://meetings.aps.org/Meeting/MAR08/Event/81519>).
17. A.N. Caruso, K.I. Pokhodnya, W.W. Shum, W.-Y. Ching, B. Anderson, M.T. Bremer, E. Vescovo, P. Rulis, A.J. Epstein, and J.S. Miller, *Finite electron Spin Polarization from the Organic-Based Magnet  $[Fe^{II}(TCNE)(NCMe)_2] [Fe^{III}Cl_4]$* , **11th International**

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18. J.B. Kortright, D.M. Lincoln, R. Shima Edelstein, and A.J. Epstein, *Bonding, Backbonding, and Spin-Polarized Molecular Orbitals: Basis for Magnetism and Semiconducting Transport in  $V(TCNE)_{x-2}$* , **11th International Conference on Molecule-based Magnets (ICMM 2008)** Florence, Italy, September 21-24, 2008.
19. J.-W. Yoo, H.W. Jang, C.B. Eom, and A.J. Epstein, *Will There Be Spin Polarized Carriers Crossing Ferromagnet/Organic Semiconductor/Ferromagnet Heterojunction?*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
20. J.L. Martin, V.N. Prigodin, and A.J. Epstein, *Low Magnetic Field Magnetoresistance Studies for Tris-(8-Hydroxyquinoline) Aluminum*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
21. K.D. Duman, N.-R. Chiou, V.N. Prigodin, and A.J. Epstein, *Temperature, Magnetic And Electric Field Dependence Of Magnetotransport for Polyaniline Nanofiber Networks*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
22. N.P. Raju, D. Lincoln, and A.J. Epstein, *Anomalous Linear Magnetoresistance (Up To 45 T) of High Temperature Organic-Based Magnetic Semiconductor  $V(Tcne)_x$  ( $x \sim 2$ )*, **2nd Topical Meeting on Spins in Organic Semiconductors (SpinOS 2009)** Salt Lake City, Utah, February 4-7, 2009.
23. M.B. Murphey, J.D. Bergeson, and A.J. Epstein, *Iron Nanoparticle Driven Spin-valve Behavior in Aligned Carbon Nanotube Arrays*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/97178>).
24. L. Nemzer, and A.J. Epstein, *An Optical Biosensing Platform using Reprecipitated Polyaniline Microparticles*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/99188>).
25. K.D. Duman, N.-R. Chiou, V.N. Prigodin, and A.J. Epstein, *Magneto-Transport in Polyaniline Nanofiber Network*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/98219>).
26. J.L. Martin, V.N. Prigodin, J.D. Bergeson, C.Y. Kao, and A.J. Epstein, *Magnetoresistance in the High Magnetic Field Regime for Organic Semiconductors*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/97177>).

27. J.-W. Yoo, H.W. Jang, C.Y. Kao, and A.J. Epstein, *Spin and Charge Injection and Transport in Ferromagnet/Organic Semiconductor/Ferromagnet Heterojunction*, **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/97173>).
28. J. Kortright, D. Lincoln, R.S. Edelstein and A.J. Epstein, *Bonding, Backbonding and Spin-Polarized Molecular Orbitals: Basis for Magnetism and Semiconducting Transport in  $V[TCNE]_{x \sim 2}$* , **Meeting of the American Physical Society**, Pittsburgh, Pennsylvania, March 16-20, 2009. (<http://meetings.aps.org/Meeting/MAR09/Event/94853>).
29. J.-W. Yoo, C.-Y. Chen, V. N. Prigodin, C. Kao, A. J. Epstein, H. W. Jang, C. W. Bark, and C. B. Eom, *Spintronic Applications of the Organic Semiconductors*, **Meeting of the American Physical Society**, Portland, Oregon, March 15-19, 2010. (<http://meetings.aps.org/Meeting/MAR10/Event/119117>).
30. B. Li, J.W. Yoo, C-Y. Kao, H.W. Jang, C.B. Eom, and A.J. Epstein, *A Multifunctional Organic Bistable Device*, **Meeting of the American Physical Society**, Portland, Oregon, March 15-19, 2010. (<http://meetings.aps.org/Meeting/MAR10/Event/123675>).
31. J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein, *An Organic-based Magnetic/non-magnetic Semiconductor as a Spin Polarized Carrier Source/channel*, **Electronic Material Conference 2010**, Notre Dame, Indiana, June 24, 2010.

#### Seminars and Colloquia, Arthur J. Epstein

1. A.J. Epstein, *Organic-based Magnets: From 'Only Worth a Footnote' to Spintronics, Photoinduced Magnetism, and Beyond*, **Colloquium, City College of New York**, New York, New York, March 22, 2006.
2. A.J. Epstein, *Organic-Based Magnets and Spintronics*, **University of California at San Diego**, San Diego, California, May 4, 2006.
3. A.J. Epstein, *Organic-Based Magnets and Spintronics*, **Colloquium, Chinese Academy of Sciences, Institute of Chemistry**, Beijing, PR China, October 12, 2006
4. A.J. Epstein, *Fabulous Polymers- from Superhydrophobicity to Magnetism*, **Faculty Lecture, 46<sup>th</sup> William Lloyd Evans Lecture, Department of Chemistry, Ohio State University**, Columbus, Ohio November 15, 2007.
5. A.J. Epstein, *Organic-based Magnets and Organic-based Spintonics*, **Colloquium, Korea University**, Seoul, Korea, October 21, 2008.

6. J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities*, **Seminar at Korea Institute of Science and Technology**, Seoul, Korea, April 21, 2010.
7. J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein, *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities*, **Seminar at Sungkyunkwan University**, Suwon, Korea, April 25, 2010.
8. J.-W. Yoo, C.-Y. Chen, V.N. Prigodin, H.W. Jang, C.W. Bark, C.B. Eom, and A.J. Epstein *Molecule-based Magnet and Organic-based Spintronics: New Science and Opportunities*, **Seminar at Korea University**, Seoul, Korea, April 27 (2010).
9. V.N. Prigodin and A.J. Epstein, *Organic Spintronics*, **Colloquium, Ioffe Institute**, St. Petersburg, Russia, May 21, 2010.

#### **VII b. Consultative and Advisory Functions**

A.J. Epstein. **Program Review of Los Alamos National Laboratory Research Program** Oct. 2007  
 Councilor, American Physical Society's Division of Condensed Matter Physics, 2008-2012

#### **VII c. Technology Assists, Transitions, and Transfers.**

A.J. Epstein: Transition of THz Detection via films of organic ferrimagnet V[TCNE]<sub>x</sub>.  
 Epstein Patents optioned by Ohio State University to Traycer Detectors Co., Columbus, OH, for development of reliable sensitive means of detecting THz and high GHz radiation to enable scanning to detect weapons and also to tumors without use of amplifiers.

### **VIII. NEW DISCOVERIES, INVENTIONS, OR PATENT DISCLOSURES**

A..J. Epstein's group discovered that the 2D layered organic-based magnet [Fe(TCNE)(NCMe)<sub>2</sub>][FeCl<sub>4</sub>] (TCNE = tetracyanoethylene) synthesized in the labs of Joel Miller exhibits a unique magnetic bifurcation between the field-cooled and zero-field-cooled states, which cannot be explained by either superparamagnetic behavior or spin freezing due to spin glass order. This irreversible magnetic bistability is described through consideration of the ensemble of uncoupled 2D Ising layers and their magnetization reversal initiated by a field-induced bubble formation ("*bubble bursting*") in individual layers. The bubble nucleation rate strongly depends on the external field and temperature resulting in anomalous magnetic relaxation. The "burst of bubbles" results in a rapid change in magnetic permeability in a narrow range of magnetic fields. Following is an article describing this phenomenon: J.-W. Yoo, V.N. Prigodin, W.W. Shum, K.I. Pokhodnya, J.S. Miller, and A.J. Epstein, *Magnetic Bistability and Nucleation of Magnetic Bubbles in Layered 2D Organic-based Magnet [Fe(TCNE)(NCMe)<sub>2</sub>][FeCl<sub>4</sub>]*, **Physical Review Letters** **101**, 197206/1-4 (2008).

## IX. HONORS/AWARDS

### Arthur J. Epstein Selected Honors/Awards, during this grant period

Plenary Lecturer, Int. Conf. on Science and Technology of Synth. Metals, Dublin, Ireland, July 2006

American Physical Society's James C. McGroddy Prize for New Materials (2007) for discovery of organic-based magnets and elucidating their properties

Councilor, American Physical Society's Division of Condensed Matter Physics, 2008-2012

### Arthur J. Epstein Selected Lifetime Honors/Awards

Fellow, American Physical Society, 1981

Adjunct Professor of Physics, University of Florida, Gainesville, Florida, 1983-87

Frontiers of Science Lecturer, University of Florida, Gainesville, Florida, October 1984; October 1996

“Hottest” Paper in Chemistry, Institute of Scientific Information, Jan.-Feb., 1990

Plenary Lecturer, Int. Conf. on Science and Technology of Synthetic Metals, Seoul, Korea, July 1994

Plenary Lecturer, The Fifth International Conference on Molecule-Based Magnets, Osaka, Japan, July 1996

Adjunct Professor of Chemistry, University of Utah, Salt Lake City, Utah, 1997-present

Lecturer, Weissberger Williams Lecture Series, Eastman Kodak Company, Rochester, NY, 9 Dec. 1997

Visiting Lecturer of the Chemistry Research Promotion Center, Taipei, Taiwan, ROC, January 2000.

Guest Professor, November 2001. Hosted by University of Tokyo, Wasada University, Tokyo Metropolitan University, Kyoto University and Osaka University

University Distinguished Lecturer 2001-2002, The Ohio State University

Watkins Visiting Professor, Wichita State University, March 4-6, 2002.

Lecturer, Center for Molecular Science Forum, Beijing, PRC, 16 July 2002

Honorary Professor, Institute of Chemistry of the Chinese Academy of Sciences, Beijing, PRC, 2002

Honorary Professor, Jilin University, Changchun, PRC, 2002

Fellow, American Association for the Advancement of Science, 2002

Highly Cited Researcher in Physics, ISI, 2002

X Symposium Lecturer, MRS Fall Meeting, 2002

William Fowler Award for Distinguished Work in Physics, Ohio Section American Phys. Soc., 2003

Plenary Lecturer, Int. Conf. on Science and Technology of Synth. Metals, Wollongong, Australia, June 2004